BARIUM STRONTIUM TITANATE THIN FILMS BY OFF-AXIS CO-SPUTTERING AND INVERTED CYLINDRICAL MAGNETRON SPUTTERING

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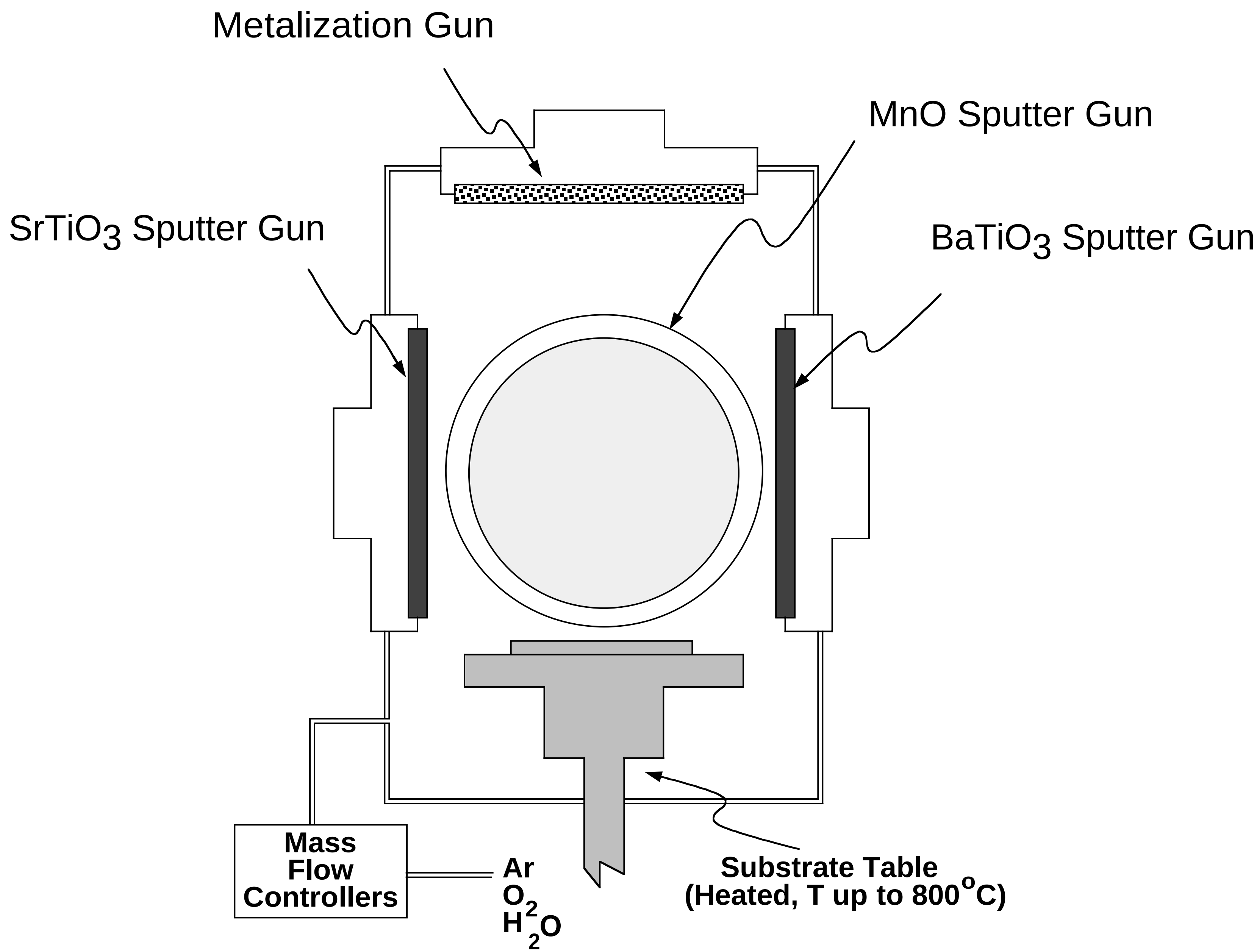
ABSTRACT

Thin films of $Ba_{(1-x)}Sr_xTiO_3$ (BST) have been deposited by off-axis co-sputtering targets of $SrTiO_3$ (STO) and $BaTiO_3$ (BTO). The films were grown on (100) MgO and $LaAlO_3$ substrates for substrate temperatures between 550 to 800° C. $Ba_{0.5}Sr_{0.5}TiO_3$ films were also deposited by Inverted Cylindrical Magnetron (ICM) sputtering. The O_2/Ar sputtering gas was 150 µm for the off-axis co-sputtering and 400 µm for the ICM sputtering. For the off-axis technique, the film composition was set by the relative target power levels. X-ray diffractometry indicated the as-deposited BST films were predominantly (h00) oriented, however, mixed orientations were observed for the different deposition conditions. Post annealing the films in flowing O_2 at 780 °C resulted in greater diffraction peak heights. Capacitance-voltage characteristics at 10 GHz yielded Q's of 65 with nearly 14% tuning for BST on LAO and Q > 1000 with a 2% tuning for films on MgO. For ICM sputtered BST films the Q > 600 with tuning nearly 7% for the best films.

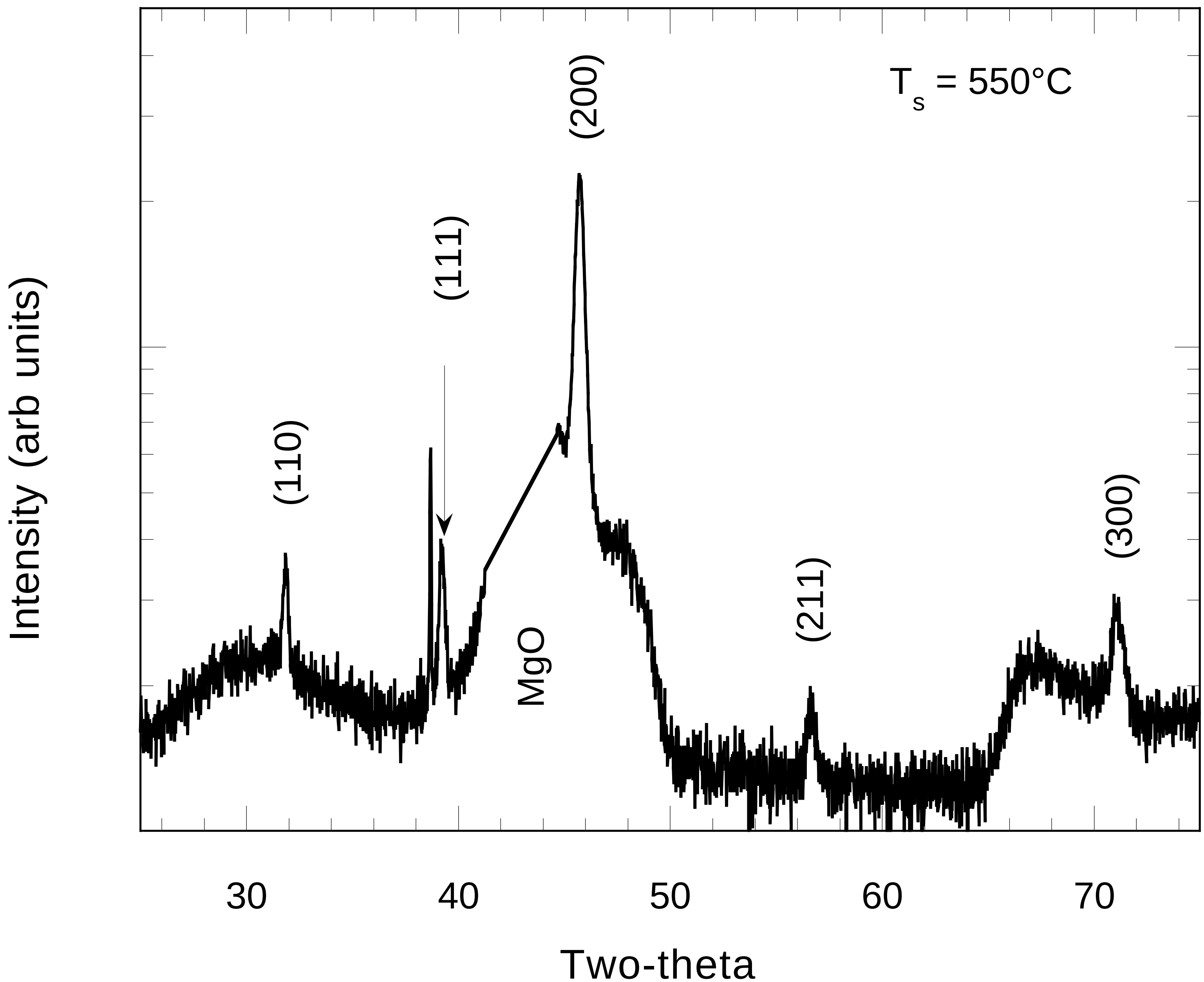
Off-Axis Co-Sputtering $BaTiO_3$ and $SrTiO_3$ to form $Ba_{(1-x)}Sr_xTiO_3$ Thin Films

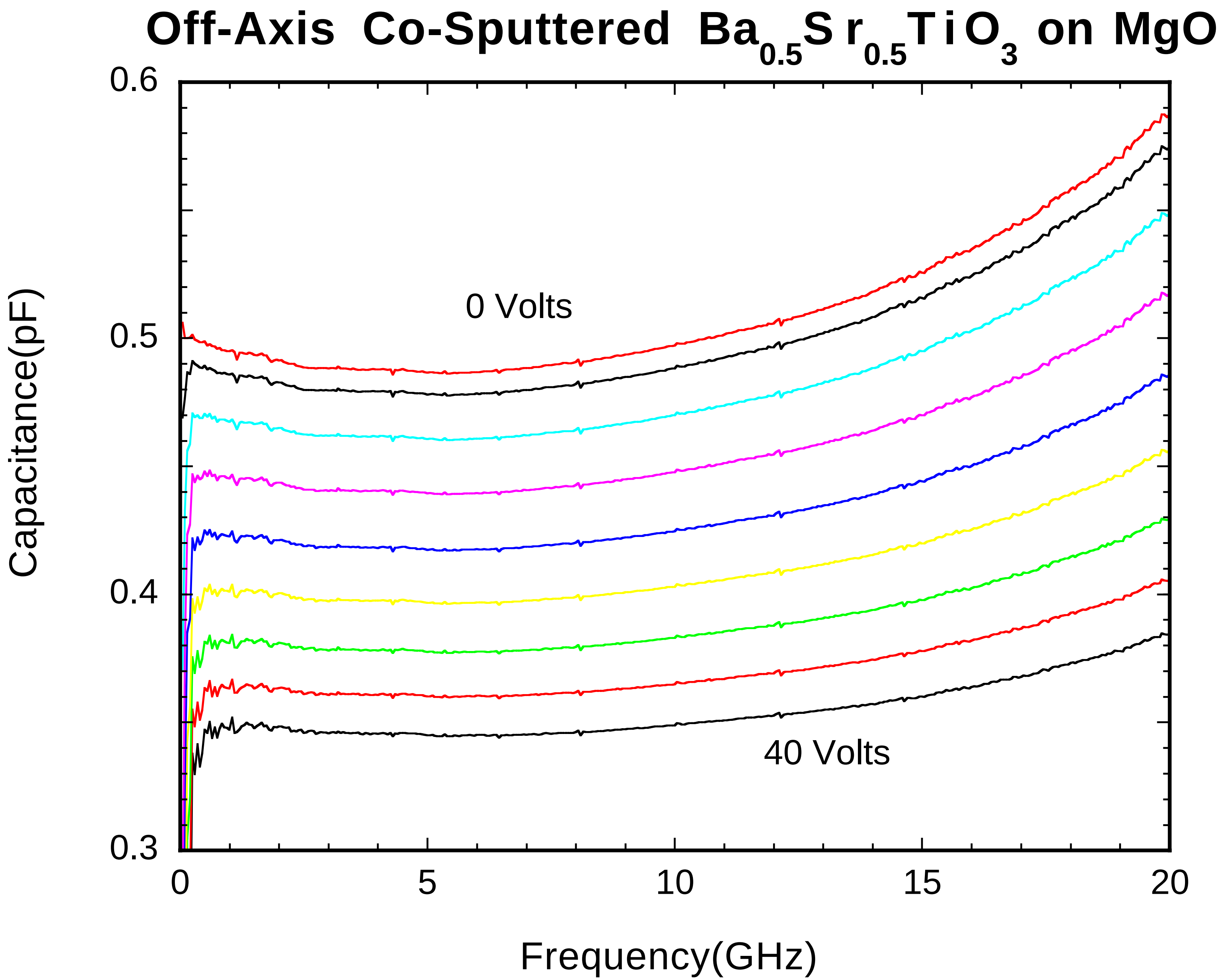
Thin films of $Ba_{(1-x)}Sr_xTiO_3$ (BST) were deposited by an off-axis co-sputtering of facing targets of $BaTiO_3$ (BTO) and $SrTiO_3$ (STO). The facing targets geometry lends itself for the reaction of BTO and STO in the plasma at the opposite target surface forming BST. Additionally, reaction of the constituents of each target may react on the substrate surface or during the post anneal at 780° C. X-ray diffraction of as-deposited films showed the existence of the BST structure indicating that the BST reaction occurs predominantly in the plasma or on the substrate surface. Post annealing serves to reduce defects and increase grain size. The geometry is such that negative ions are not accelerated towards the substrate and thus resputtering of the growing film is minimized. Additionally, the high sputtering gas pressure (150 μm) serves to thermalize all constituents and further reduces any resputtering effects at the substrate. The low temperature deposition (550° C) produces films having reasonable Q and tuning. The trade-off between Q and tuning is under investigation in relation to the structural properties of the films. As the substrate temperature was increased, the surface became rough in relation to the specular surface of films deposited at 550° C. The addition of a third off-axis magnetron gun permits doping the film with Mn to any level by setting the power level. This will allow for very small (<<1%) to very high doping from one target. The low deposition rate (~500 Å/h) inherent to off-axis sputtering allows for a well ordered film on the substrate which is ideal for thin films as used in parallel plate capacitors.

Off-Axis Co-Sputtering System

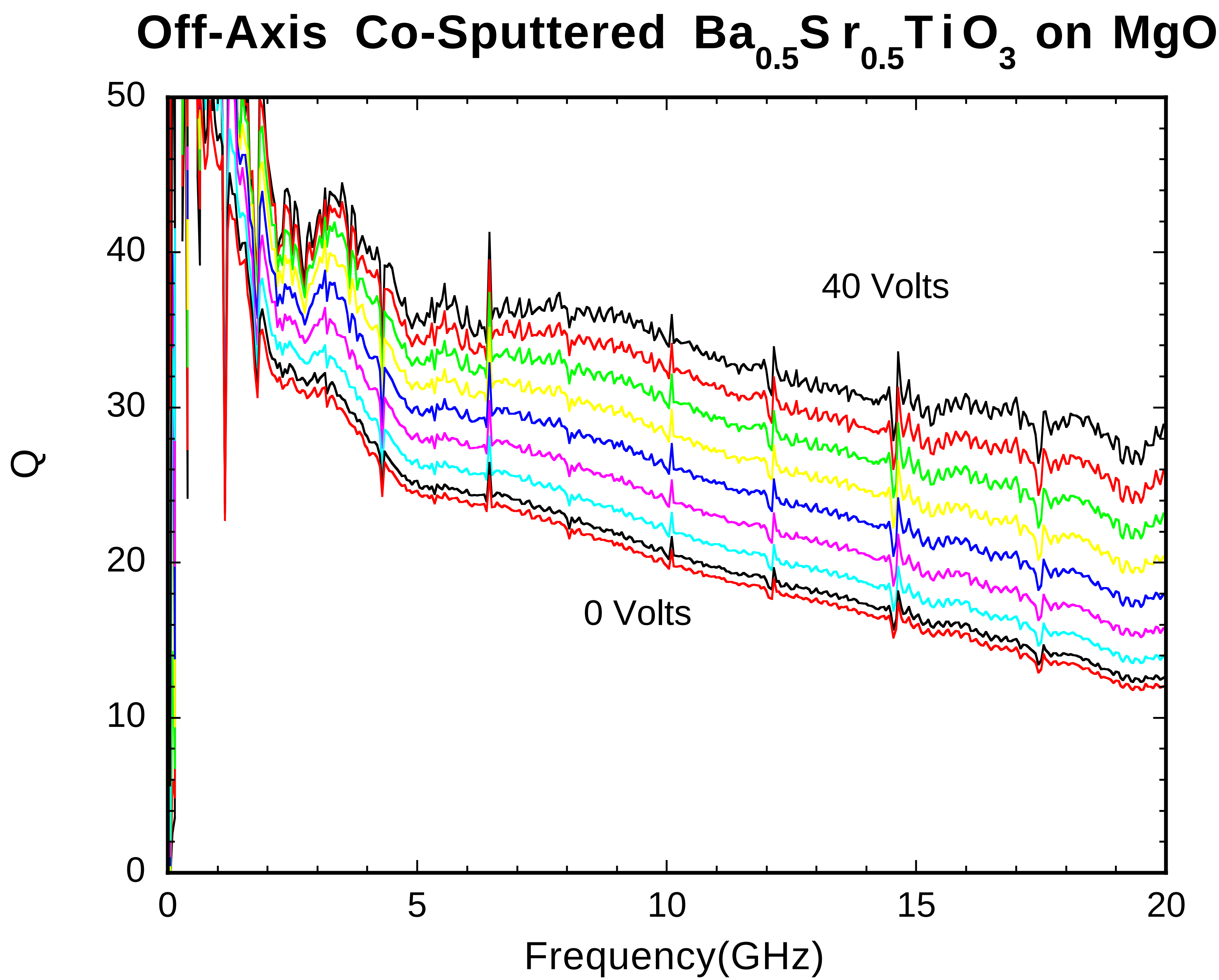


Off-Axis Co-Sputtered $Ba_{0.5}Sr_{0.5}TiO_3$ on MgO

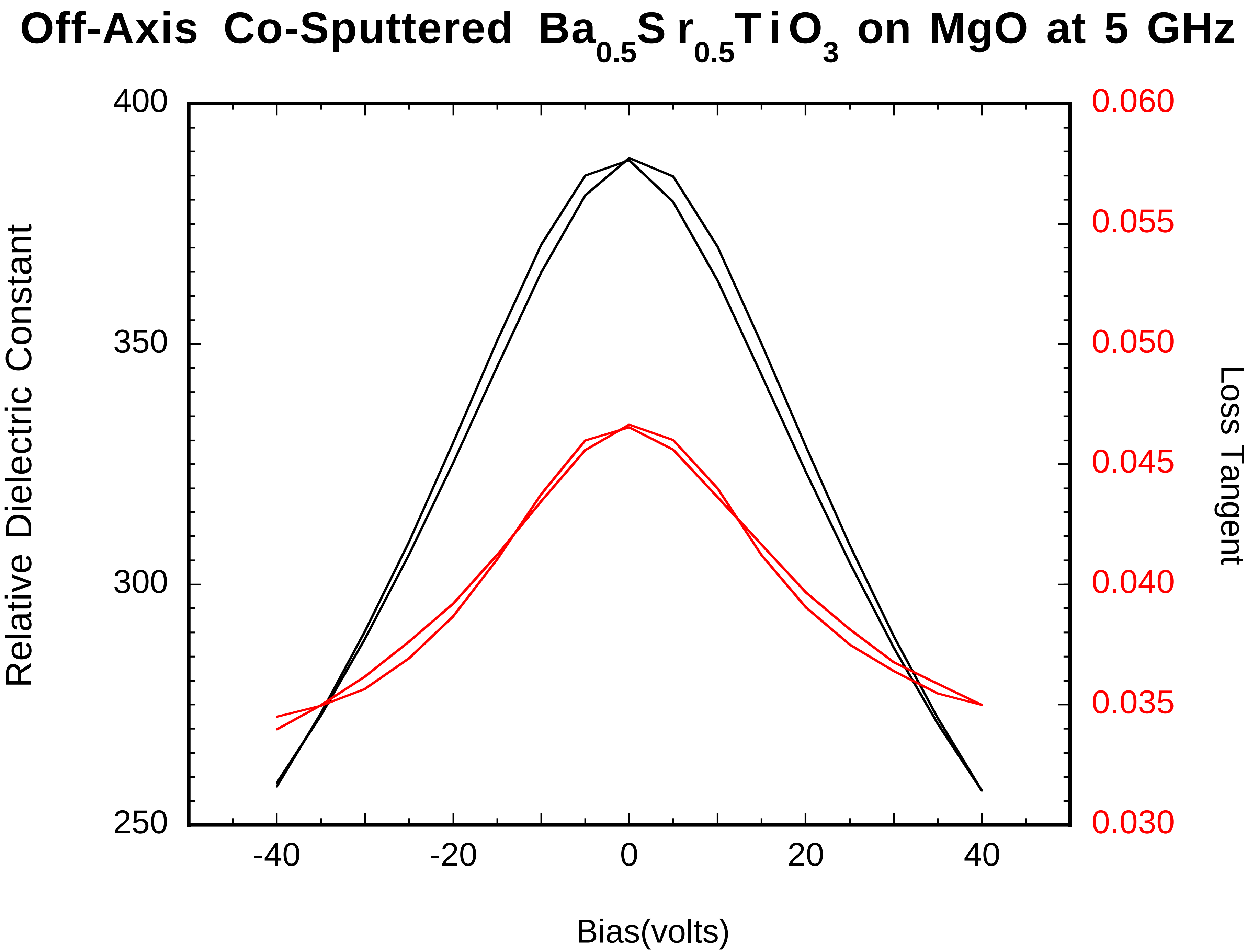




Off-Axis Co-Sputtered $Ba_{0.5}Sr_{0.5}TiO_3$ on MgO
Capacitance(pF)
0.6
0.5
0.4
0.3
0 Volts
40 Volts
0
5
10
15
20
Frequency(GHz)



Off-Axis Co-Sputtered $Ba_{0.5}Sr_{0.5}TiO_3$ on MgO
Q
Frequency(GHz)
40 Volts
0 Volts
0
10
20
30
40
50
0
5
10
15
20

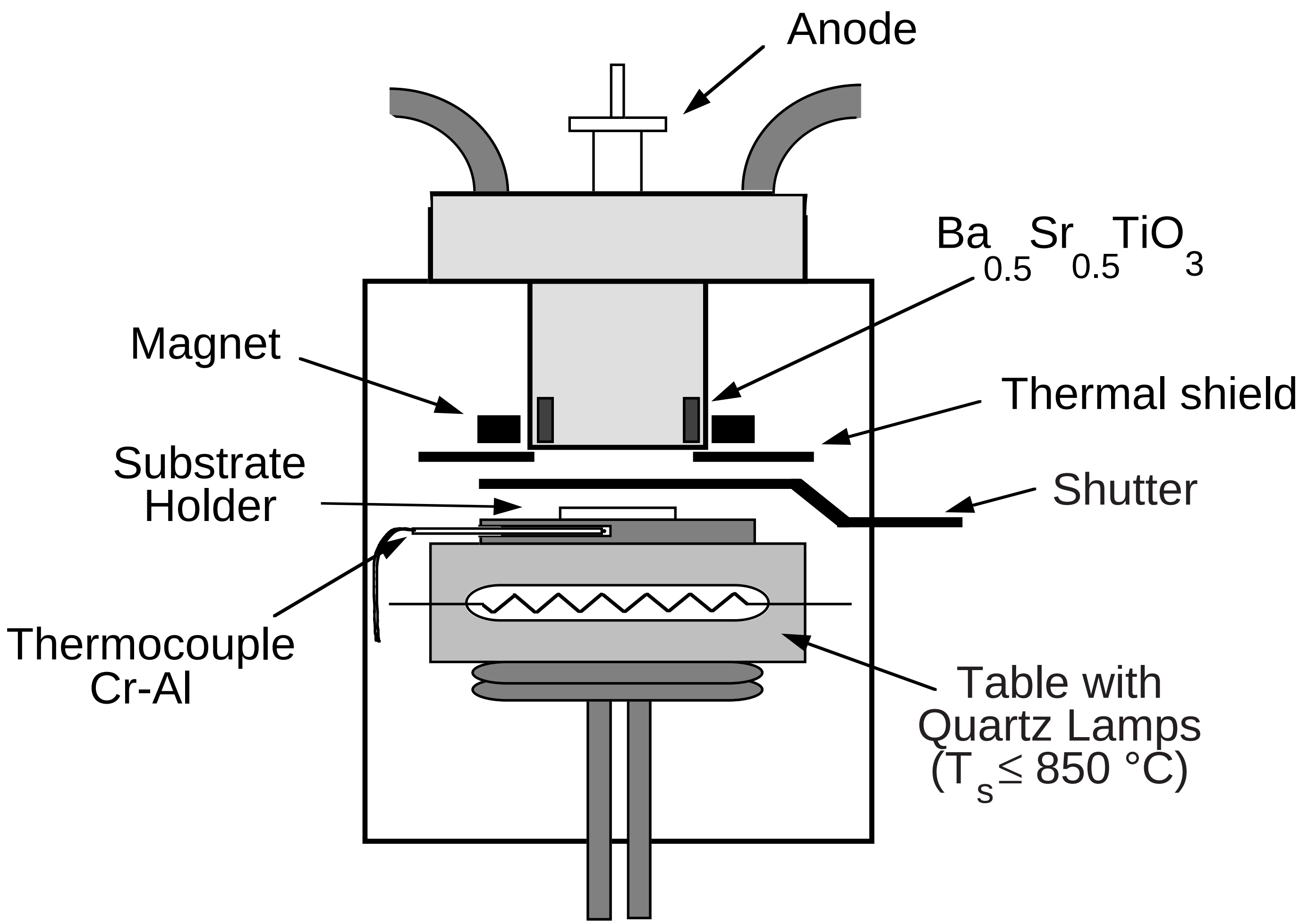


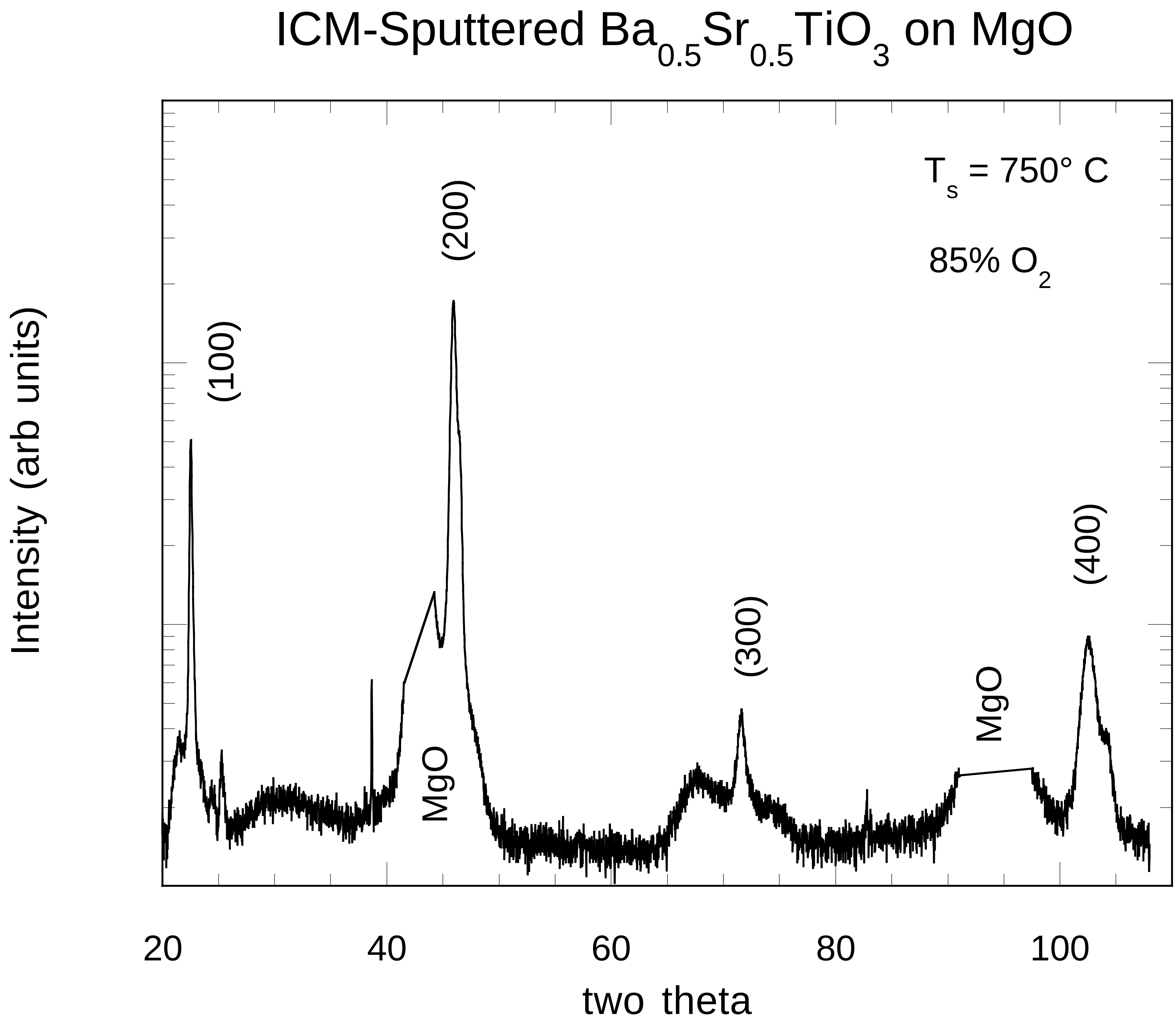
Off-Axis Co-Sputtered $Ba_{0.5}Sr_{0.5}TiO_3$ on MgO at 5 GHz
400
350
300
250
0.060
0.055
0.050
0.045
0.040
0.035
0.030
-40
-20
0
20
40
Relative Dielectric Constant
Loss Tangent
Bias(volts)

Inverted Cylindrical Magnetron Sputtering of $Ba_{0.5}Sr_{0.5}TiO_3$ Thin Films

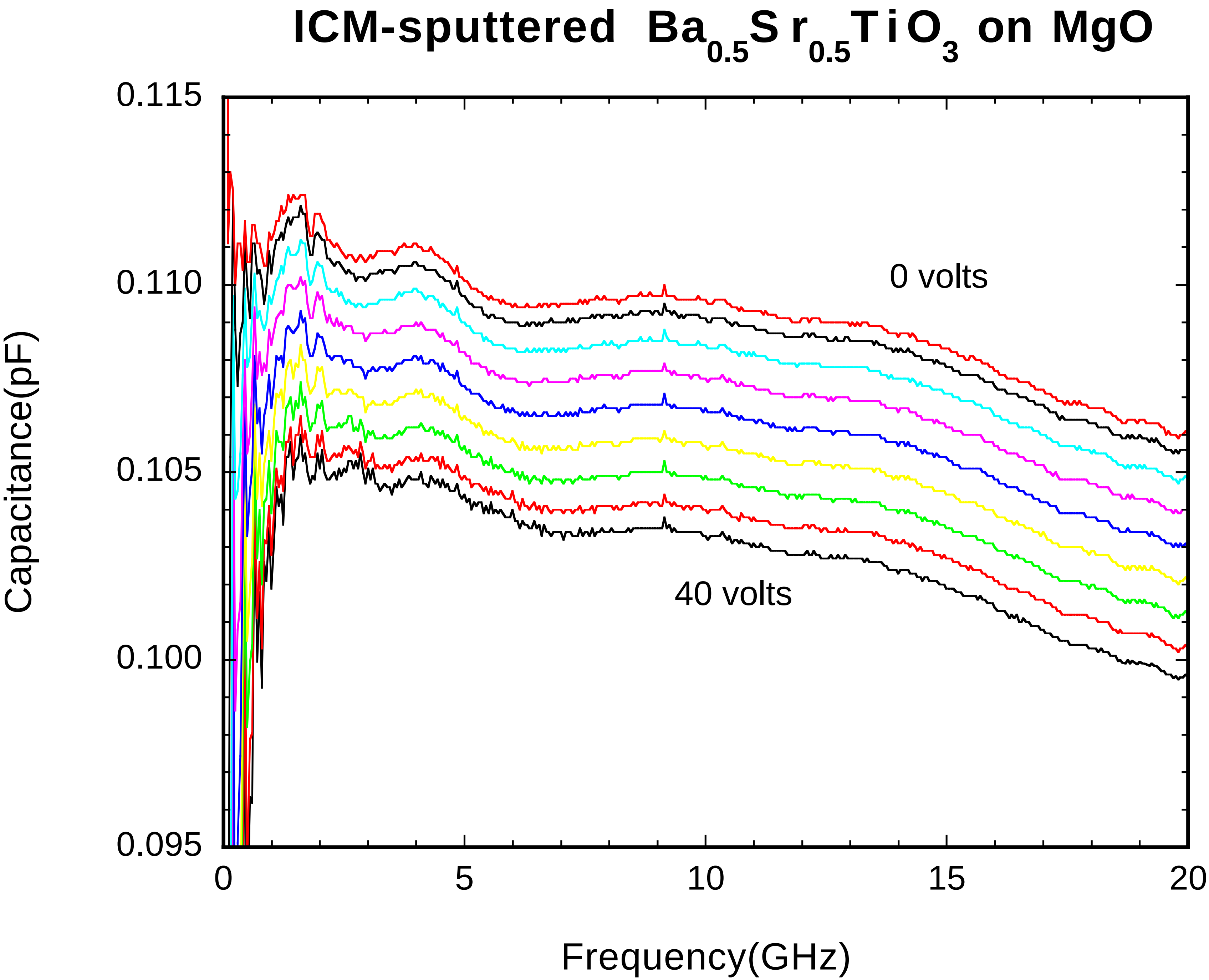
Thin films of $Ba_{0.5}Sr_{0.5}TiO_3$ have been sputter deposited by Inverted Cylindrical Magnetron (ICM) sputtering for several substrate temperatures, oxygen partial pressures, and substrate-to-target spacings. This technique uses a single $Ba_{0.5}Sr_{0.5}TiO_3$ composite target, high sputtering gas pressure (400 µm), and substrate temperatures ranging from 550° C to 800° C. The deposition rate is relatively high at 2 - 3 kÅ/h. At the lower substrate temperatures, the films were specular to the eye becoming increasing rough for higher substrate temperatures. Independent of the surface roughness, some films had very high Q, values exceeding the resolution of the device/instrumentation. The preferred orientation of the films was very dependent on the sputtering parameters of temperature, oxygen partial pressure, target-to-substrate spacing, and substrate type. Oxygen partial pressure ranging from 2.5% to 100% were investigated. Films made with little oxygen did not have the BST structure. The optimum oxygen partial pressure was observed to be approximately 85%. The high sputtering gas pressure and the ICM gun geometry virtually eliminate the adverse effects of negative ion bombardment which is inherent to conventional parallel plate sputtering of oxide films. Using ICM sputtering, we have produced BST thin films having very high Q, beyond the resolution of the device/instrument with reasonable tuning of nearly 7%.

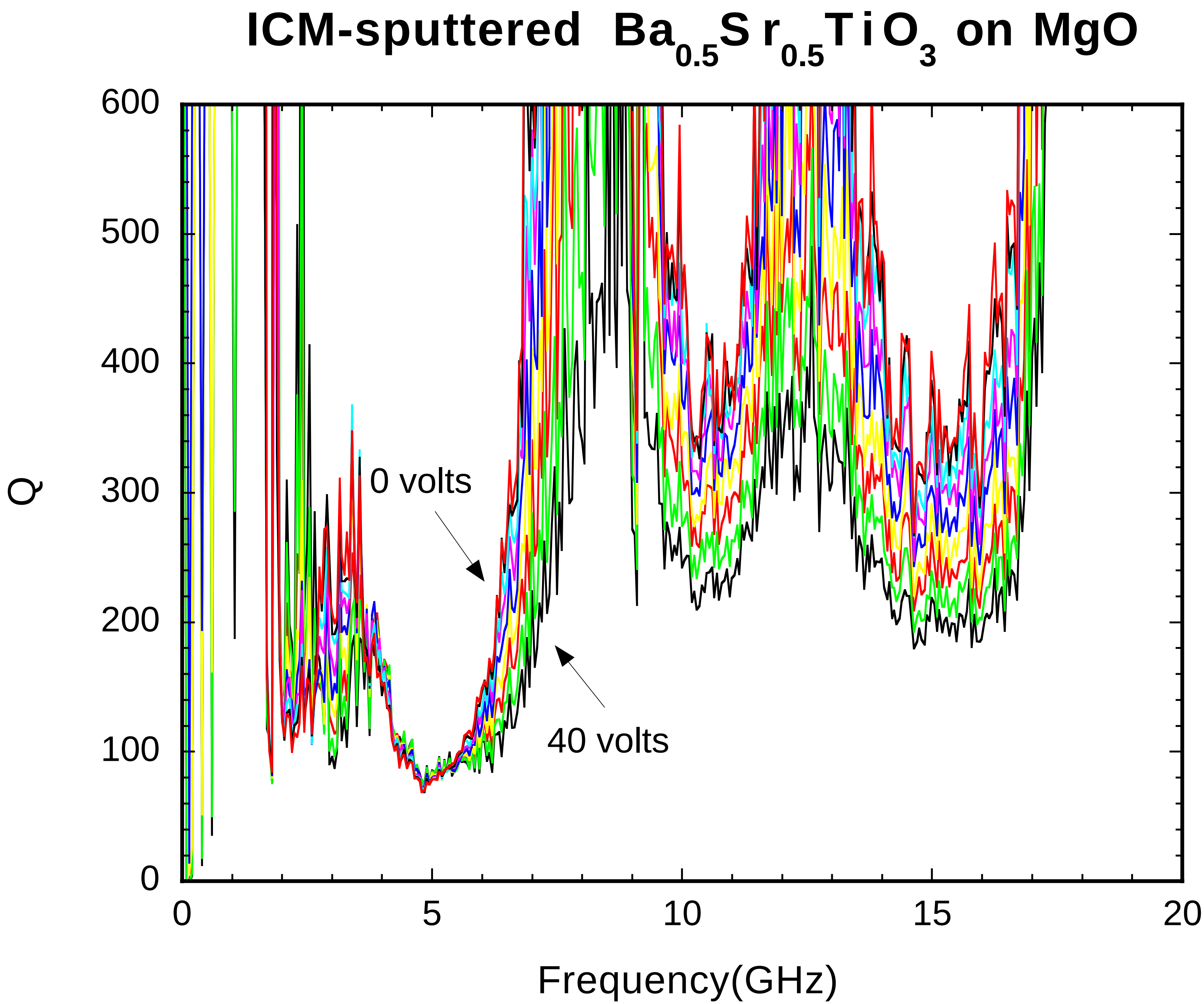
Inverted Cylindrical Magnetron Sputtering System

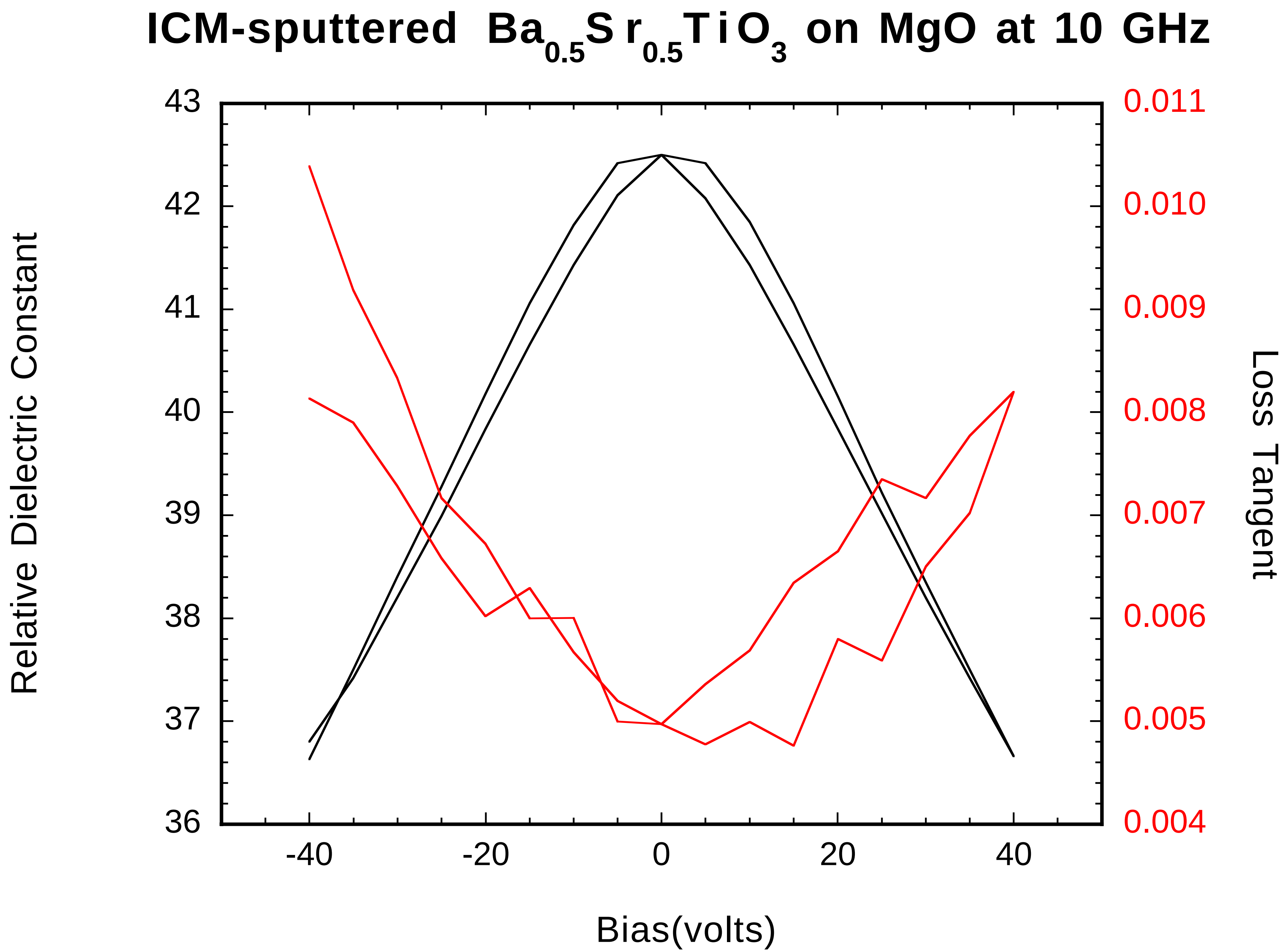




ICM-Sputtered Ba0.5Sr0.5TiO3 on MgO
Ts = 750° C
85% O2
(100)
(200)
(300)
(400)
MgO
MgO
Intensity (arb units)
20
40
60
80
100
two theta


ICM-sputtered $Ba_{0.5}Sr_{0.5}TiO_3$ on MgO
0.115
0.110
0.105
0.100
0.095
0
5
10
15
20
Capacitance(pF)
Frequency(GHz)
0 volts
40 volts


ICM-sputtered $Ba_{0.5}Sr_{0.5}TiO_3$ on MgO
Q
600
500
400
300
200
100
0
0 volts
40 volts
0
5
10
15
20
Frequency(GHz)



ICM-sputtered $Ba_{0.5}Sr_{0.5}TiO_3$ on MgO at 10 GHz
Relative Dielectric Constant
Loss Tangent
Bias(volts)
43
42
41
40
39
38
37
36
0.011
0.010
0.009
0.008
0.007
0.006
0.005
0.004
-40
-20
0
20
40

Characteristics of Unconventional Sputter Deposition Techniques for $Ba_{(1-x)}Sr_xTiO_3$ Thin Films

Inverted Cylindrical Magnetron Sputtering (ICM)	**Off-Axis Co-Sputtering $BaTiO_3$ and $SrTiO_3$**
High rate: 2-3 kÅ/h	Slow growth rate: to 500 Å/h
Ideal for oxide films > 1000Å in thickness	Ideal for thin oxide films < 1000 Å
Geometry and high sputtering pressure (400 µm) eliminate back sputtering	Geometry and high sputtering pressure (150 µm) eliminate back sputtering
Surface morphology dependent on deposition temperature and sputter pressure	Any Ba/Sr metals fraction using only two targets and set by relative power levels of each target.
Film orientation dependent on sputtering parameters, (h00) or mixed orientations	Low temperature (550° C) substrate temperature results in smooth surface morphology
	Ease of cation doping with third gun to any concentration
	Preferred orientation is dependent on deposition parameters, (h00) or mixed orientations

Summary of Properties for Selected BST thin Films

Technique/substrate	Q_{avg}	Tuning (%)	Quality Factor*	Remarks
OA9-MgO	250	4.93	12.3	a_0 = 3.953, T_s = 550°C, 25% O_2
OA16-LAO	68	13.8	9.4	a_0 = 3.956, T_s = 550°C, 25% O_2
OA17-MgO	780	2.1	16.4	T_s = 550°C, 25% O_2, seed layer
OA21-MgO	27	30	8.10	a_0 = 3.964, T_s = 550°C, 25% O_2, single target
OA25-MgO	59	10.4	6.1	a_0 = 3.942, T_s = 550°C, Ar/O_2/H_2O, seed layer
OA31-MgO	26.5	25.2	6.7	a_0 = 3.980, T_s = 550°C, 25% O_2, seed layer
ICM10-MgO	43.5	4.1	1.8	a_0 = 3.964, T_s = 550°C, seed layer @ 750° C
ICM17-MgO	45.5	7.1	3.2	a_0 = 3.981, T_s = 550°C, 100% O_2
ICM20-MgO	155.5	5.6	8.7	a_0 = 3.958, T_s = 750°C, 85% O_2
ICM22-MgO	>600	6.7	>40.2	a_0 = 3.954, T_s = 750°C, 85% O_2, pure (h00)
ICM24-MgO	>600	0.74	>4.4	a_0 = 3.906, T_s = 750°C, 100% O_2
ICM32-LAO	19.5	9.1	1.8	a_0 = 3.960, T_s = 700°C, 85% O_2

* Quality Factor = Q_{avg} x (% Tuning)

Conclusions

- $Ba_{(1-x)}Sr_xTiO_3$ thin films can be deposited for any Ba/Sr metals fraction by off-axis co-sputtering BTO and STO targets. The addition of any % dopant is easily accomplished with a third off-axis gun.
- The material and electrical properties are strongly dependent on the deposition conditions. Highly oriented films having low loss and good tuning have been observed for these films.
- ICM sputtering $Ba_{0.5}Sr_{0.5}TiO_3$ from a single target has yielded well oriented films having very low loss. Film orientation is strongly effected by oxygen partial pressure and target to substrate spacing. Low oxygen partial pressure does not result in the BST structure. 85% oxygen in the sputtering gas is optimum.
- The geometry and high sputtering gas pressure of the off-axis and ICM sputtering techniques essentially eliminates the adverse effects of negative ion bombardment of the growing film.
- These two unconventional sputtering techniques are suitable for the growth of BST for many microwave applications requiring low loss and reasonable tuning ferroelectric thin films.